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(54) **Biaxial orientierte Polyesterfolie, Verfahren zu ihrer Herstellung und ihre Verwendung zum Verpacken von Leben- und Genussmittel**

(57) Die Erfindung betrifft eine biaxial orientierte Polyesterfolie mit sehr guten optischen Eigenschaften und gutem Verarbeitungsverhalten, die nach ihrer Metallisierung oder nach ihrer Beschichtung mit oxidischen Materialien eine hohe Sauerstoffbarriere aufweist, aufgebaut aus mindestens einer Basisschicht B und mindestens einer auf dieser Basisschicht aufgetragenen Schicht A, wobei diese Schicht A eine definierte Anzahl von Erhebungen mit einer definierten Höhe und einem definierten Durchmesser aufweist. Die Erfindung betrifft weiterhin ein Verfahren zur Herstellung der Folie und ihre Verwendung.

EP 0 903 221 A2



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Peiffer et al.

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 (45) Date of Patent: **Mar. 4, 2003**

(54) **BIAXIALLY ORIENTED POLYESTER FILM,
 THE USE THEREOF, AND PROCESS FOR
 THE PRODUCTION THEREOF**

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 patent is extended or adjusted under 35
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This patent is subject to a terminal dis-
 claimer.

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 22, 1998, now abandoned.

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(51) Int. Cl.⁷ **B32B 3/00; B32B 15/04;
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 428/694 SC; 156/244.24; 264/290.2; 427/147**

(58) Field of Search **428/216, 141,
 428/323, 332, 339, 480, 910, 33.6, 694.57,
 694.56, 457, 458; 264/290.2; 156/244.24;
 427/146, 147**

(56) **References Cited**

U.S. PATENT DOCUMENTS

3,515,626 A 6/1970 Duffield
 3,958,064 A 5/1976 Brekken et al.

(List continued on next page.)

FOREIGN PATENT DOCUMENTS

DE 1 694 404 4/1971
 DE 22 30 970 2/1973
 DE 38 01 535 7/1988

(List continued on next page.)

OTHER PUBLICATIONS

Kimura, S.F. et al., FTIR Spectroscopic Study on Crystallization Process of Poly (ethylene-2,6-naphthalate), 35 Journal of Polymer Science: Polymer Physics 2741-2747 (1997).

(List continued on next page.)

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(57) **ABSTRACT**

The invention relates to a biaxially oriented polyester film which has very good optical properties and good processing behavior and which after it has been metallized or coated with oxidic materials exhibits a high oxygen barrier and which is built up from at least one base layer B and at least one layer A applied on top of this base layer, this layer A having a number N of elevations/protrusions per mm² of film surface area which is related to their respective heights h and diameters d by the following equations:

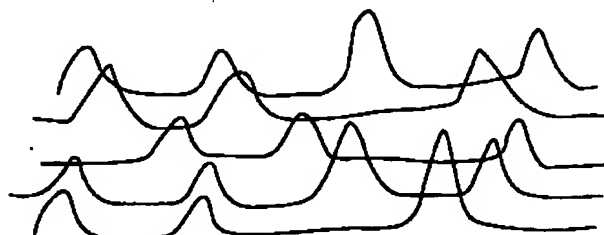
$$\log N/\text{mm}^2 < A_{\text{h}} \cdot B_{\text{d}} \cdot \log h/\mu\text{m}, 0.01 \mu\text{m} < h < 10 \mu\text{m} \quad (1)$$

wherein $A_{\text{h}}=1.4$; $B_{\text{d}}=2.5$

$$\log N/\text{mm}^2 < A_{\text{d}} \cdot B_{\text{h}} \cdot \log d/\mu\text{m}, 0.4 \mu\text{m} < d < 10 \mu\text{m} \quad (2)$$

wherein $A_{\text{d}}=3.4$; $B_{\text{h}}=2.4$.

17 Claims, 6 Drawing Sheets



OXYGEN TRANSMISSION OTR: $0.4 \text{ cm}^3/\text{m}^2 \text{ d bar}$

ROUGHNESS R_{a} : 55nm

FIGURE 1a: VERY GOOD OXYGEN BARRIER

U.S. PATENT DOCUMENTS

4,042,569 A	8/1977	Bell et al.	
4,252,885 A	2/1981	McGrail et al.	
4,399,179 A	8/1983	Minami et al.	
4,493,872 A	1/1985	Funderburk et al.	
4,615,939 A	10/1986	Corsi et al.	
4,622,237 A	11/1986	Lori	
5,236,680 A	8/1993	Nakazawa et al.	
5,236,683 A	8/1993	Nakazawa et al.	
5,242,757 A	9/1993	Buisine et al.	
5,429,785 A	7/1995	Jolliffe	
5,453,260 A	9/1995	Nakazawa et al.	
5,468,527 A	11/1995	Peiffer et al.	
5,506,014 A	4/1996	Minnick	
5,747,174 A	5/1998	Kimura et al.	
5,783,017 A	7/1998	Bosmell	
6,194,054 B1 *	2/2001	Peiffer et al.	428/141
6,214,440 B1 *	4/2001	Peiffer et al.	156/244.24

FOREIGN PATENT DOCUMENTS

DE	43 06 155	9/1994
EP	0 035 835	9/1981
EP	0 061 796	10/1982
EP	0 088 635	9/1983
EP	0 124 291	11/1984
EP	0 135 451	3/1985
EP	0 144 878	6/1985
EP	0 236 945	9/1987
EP	0 296 620	12/1988
EP	0 374 646	12/1989
EP	0 368 278 A2	5/1990
EP	0 378 154	7/1990
EP	0 378 955	7/1990
EP	0 402 861	12/1990
EP	0 490 665	6/1992
EP	0 502 745	9/1992
EP	0 514 129	11/1992
EP	0 515 096	11/1992
EP	0 580 404	1/1994
EP	0 602 964	6/1994
EP	0 604 057	6/1994
EP	0 609 060	8/1994

EP	0 612 790	8/1994
EP	0 624 454 A1	11/1994
EP	0 659 810	6/1995
EP	0 663 286	7/1995
EP	0 678 554 A1	10/1995
EP	0 685 509	12/1995
EP	0 707 979	4/1996
EP	0 826 478	3/1998
WO	WO 94/13476	6/1994
WO	WO 94/13481	6/1994
WO	WO 98/13414	4/1998
WO	WO 98/13415	4/1998
WO	WO 88/10188	12/1998

OTHER PUBLICATIONS

Bursch et al., "Biaxially Oriented Polyester Film for SMD Condensers—Made by Extrusion Followed by Biaxial Stretching and Heat Fixing to Give Specified Shrinkage Parameters in the Machine Direction and Transverse Direction," Derwent Abstract, WPI Acc. No. 98-194495/199818 (1998).

Database WPI, Section Ch, Week 9615, Derwent Publications Ltd., London, GB; Class A23, An 96-148338, XP002114377 & JP 08 03679 A (Toray Indus., Inc.) (Feb. 6, 1996).

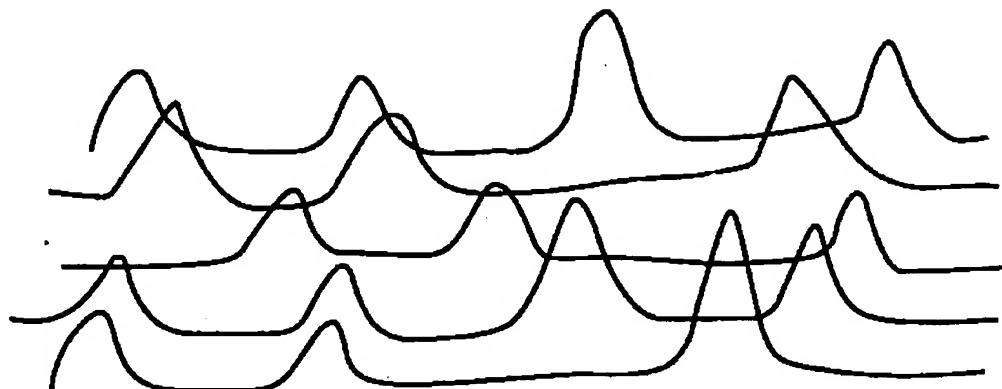
Database WPI, Section Ch, Week 9522, Derwent Publications Ltd., London, GB; Class A32, AN 95-166886, CP002114408 & JP 07 088592 A (Toray Indus., Inc.) (Apr. 4, 1995).

Database WPI, Section Ch, Week 199732, Derwent Publications Ltd., London, GB; Class A23, AN 1997-346220, XP002143058 & JP 09 141797 A (Toray Ind Inc), Jun. 3, 1997.

J. Weiss, "Parameters that Influence the Barrier Properties of Metallized Polyester and Polypropylene Films", Thin Solid Films 204; pp. 203-216 (1991).

Schmitz, Peter et al., "Films." Ullmann's Encyclopedia of Industrial Chemistry, 5th Ed., vol. A11, 1988, pp. 85-89 108-110.*

* cited by examiner

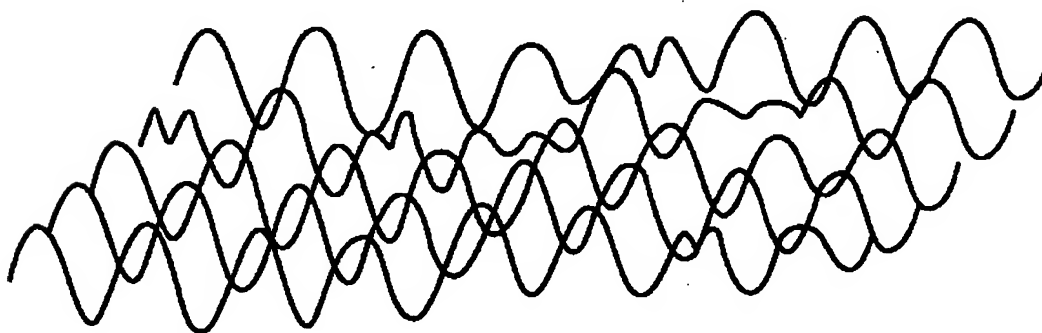


OXYGEN TRANSMISSION OTR: $0.4 \text{ cm}^3/\text{m}^2 \text{ d bar}$

ROUGHNESS R_a : 55nm

FIGURE 1a: VERY GOOD OXYGEN BARRIER

FIG. 1a



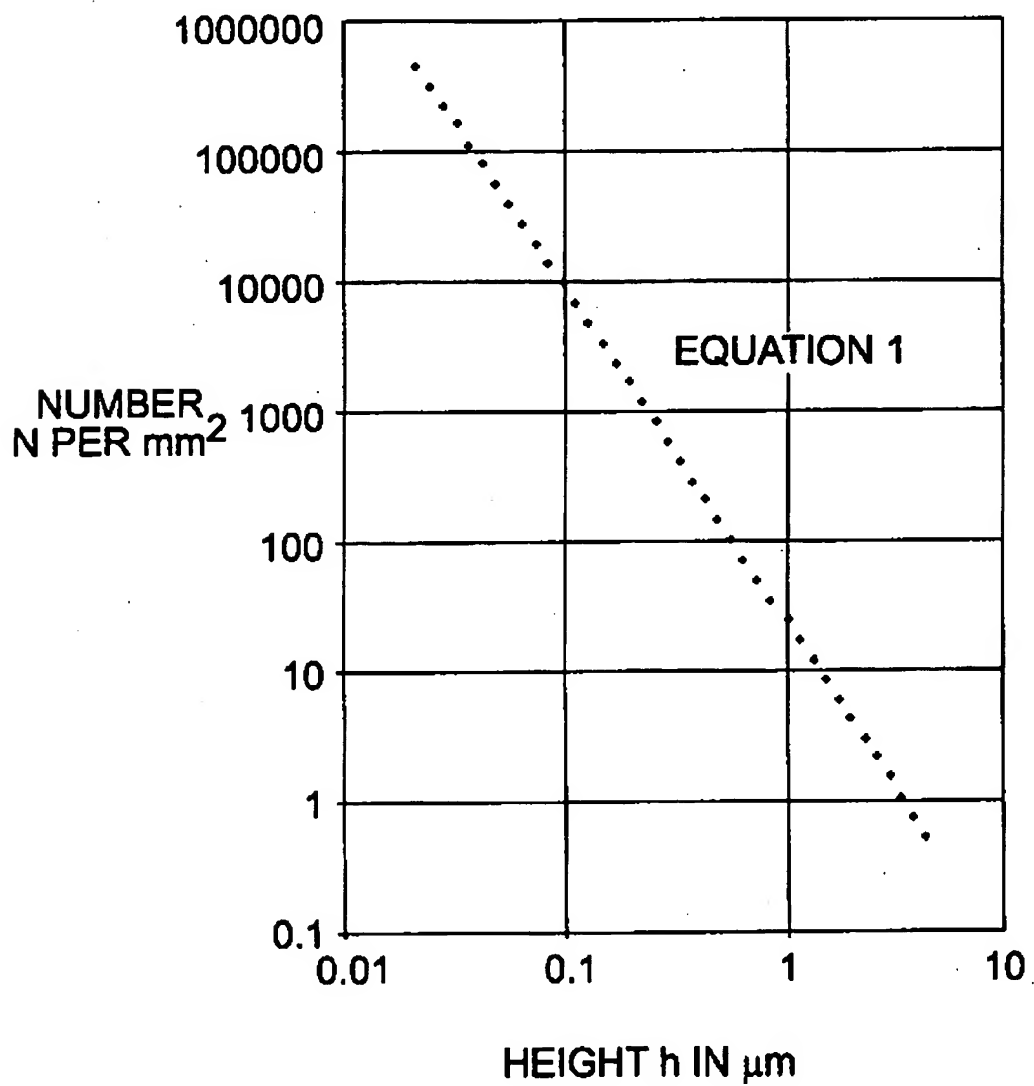
OXYGEN TRANSMISSION OTR: $3.2 \text{ cm}^3/\text{m}^2 \text{ d bar}$

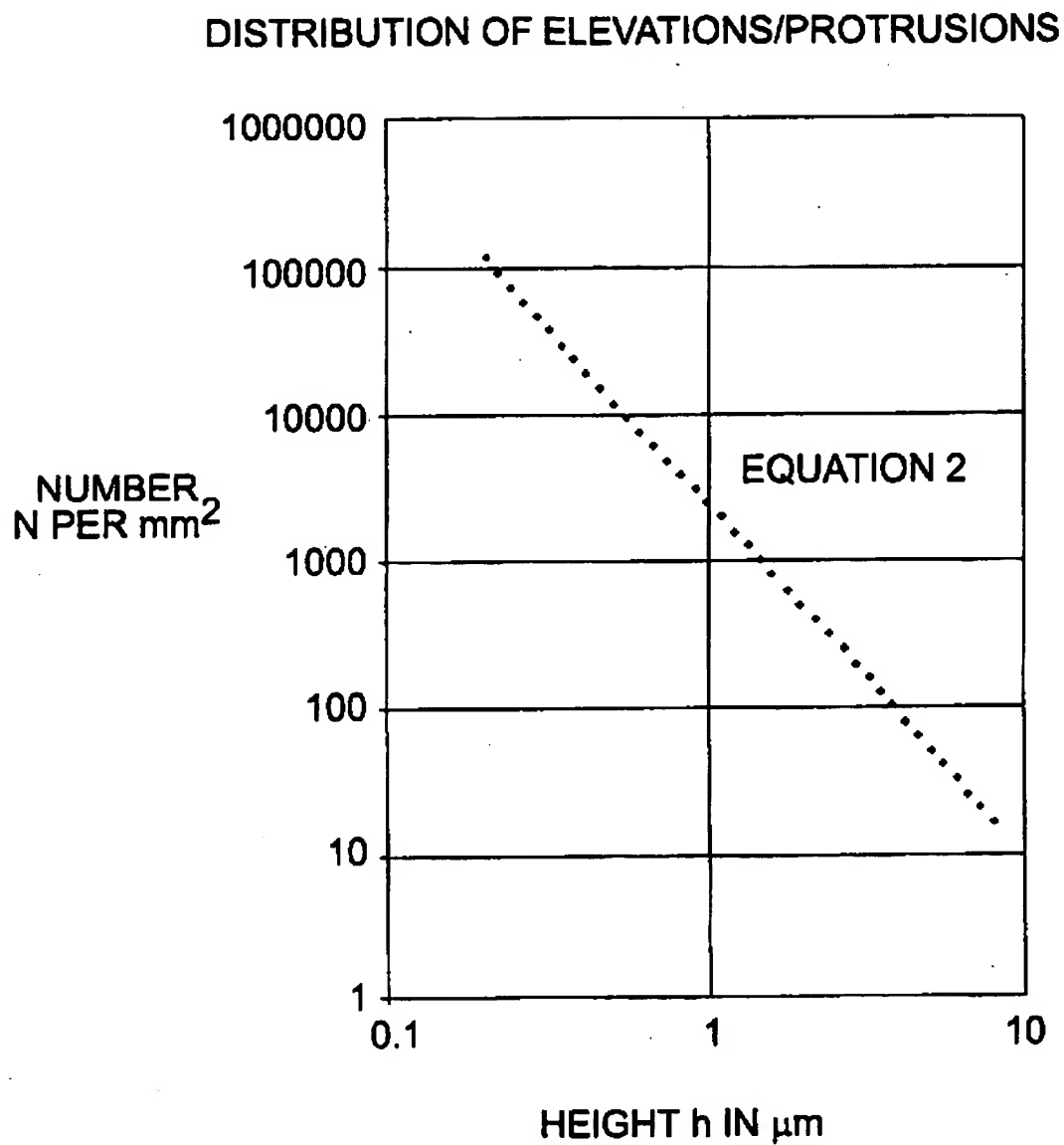
ROUGHNESS R_a : 8nm

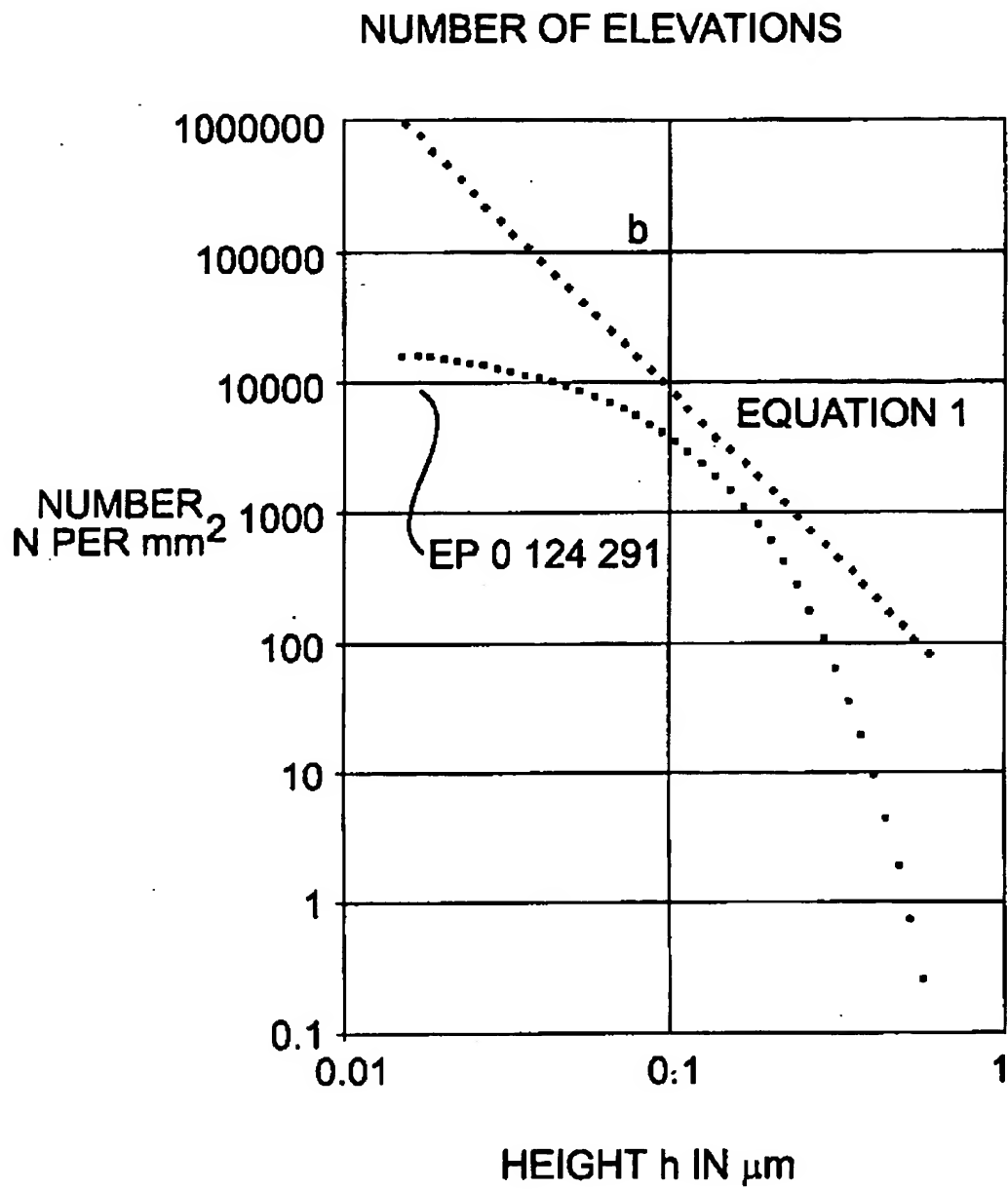
FIGURE 1b: POOR OXYGEN BARRIER

FIG. 1b

DISTRIBUTION OF ELEVATIONS/PROTRUSIONS

**FIG. 2a**

**FIG. 2b**

**FIG. 3**

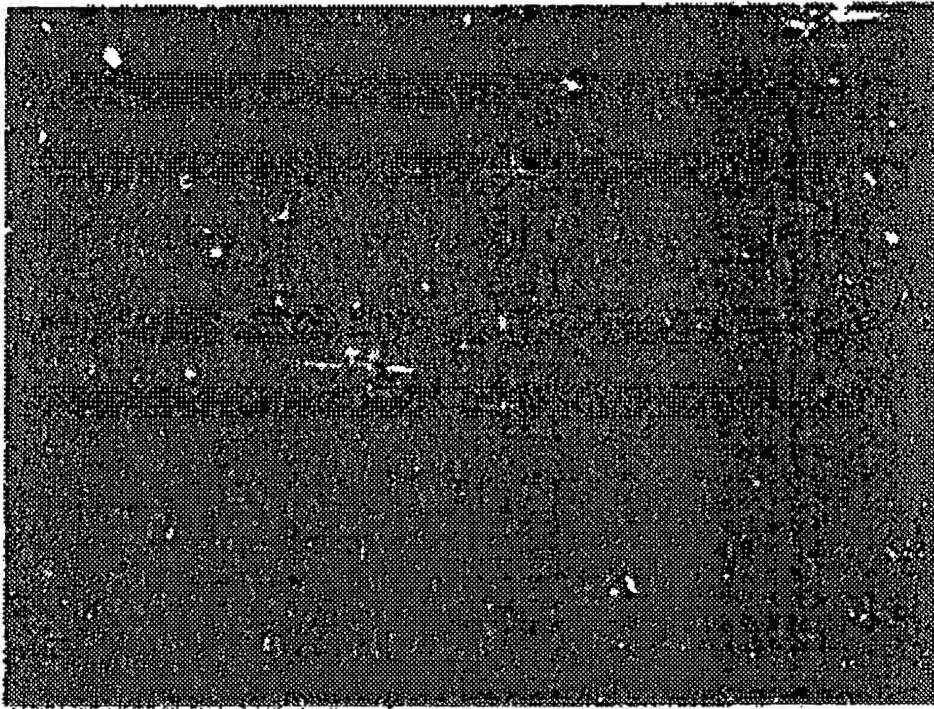
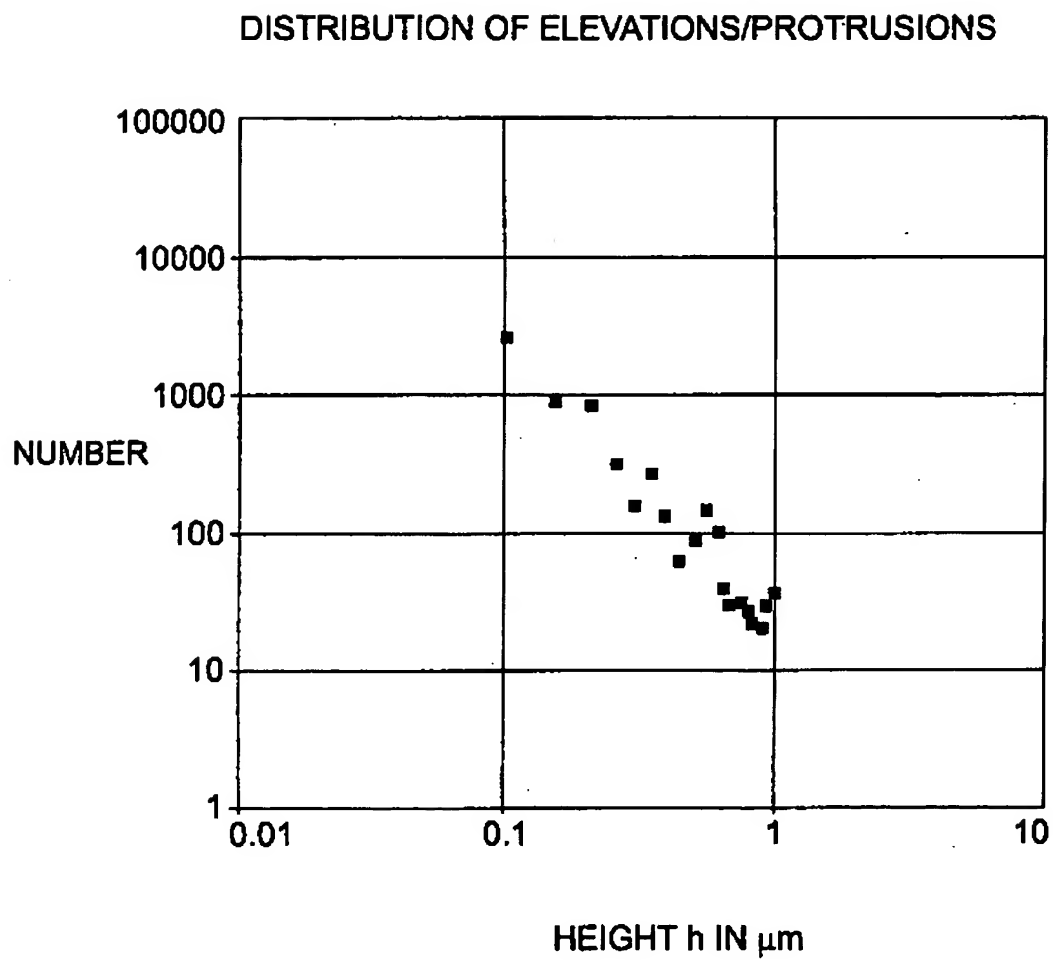


FIG. 4

**FIG. 5**

BIAXIALLY ORIENTED POLYESTER FILM, THE USE THEREOF, AND PROCESS FOR THE PRODUCTION THEREOF

This application is a continuation of Ser. No. 09/158,454 filed Sep. 22, 1998 now abandoned.

BACKGROUND OF THE INVENTION

1. Field of the Invention

Biaxially oriented polyester films are used in packaging and in industry primarily where there is a need for their advantageous properties, i.e. good optical properties, high mechanical strengths, good barrier effect, in particular against gases, good dimensional stability when heated and excellent layflat.

For most applications it is also desirable, e.g. for reasons of effective presentation, to improve the optical properties of the polyester films, in particular the gloss and the film haze, while retaining good processability of the film. It is likewise desirable to improve the barrier properties of polyester films, e.g. in order to gain new applications.

2. Description of the Related Art

The prior art has demonstrated how it is possible to improve the optical properties, in particular the gloss and the haze, of biaxially oriented polyester films.

EP-A-0 514 129 describes a transparent multilayer film which comprises a substrate of a primary layer of polymer material which, at least on one of its surfaces, has a secondary layer of polymer material which has certain concentrations and certain size distributions of glass beads and silica particles. The secondary layer can be arranged on one or on both sides of the primary layer substrate. The haze and processing properties are improved in the film, but no improvement in the gloss and the barrier properties of the film are provided in the text. There is also no indication of any kind in the text as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

EP 0 604 057 describes a transparent multilayer film which comprises a substrate of a primary layer of polymer material which is essentially free from fillers and which, at least on one of its surfaces, has a secondary layer of polymer material which contains, as filler, in a concentration of from 100 to 1000 ppm, silicone resin having an average particle diameter of from 1.5 to 12.5 μm . A disadvantage of the silicone particles is that these are comparatively expensive and do not provide an acceptable solution for the packaging market. In addition, films which are provided with pigments of this type tend to telescope more easily during reeling. In this text there is likewise no indication of any type as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

In many foodstuff packaging applications, there is demand for a high barrier effect against gases, steam and flavors (this having the same significance as low transmission or low permeability). A well known process for producing packaging of this type consists in high-vacuum aluminum metallizing of the plastic films used. Other well known processes consist in coating the films with oxidic materials (e.g. SiO_2 or Al_2O_3). Essentially, the coatings used are transparent.

The barrier effect against the substances mentioned above depends essentially on the type of the polymers in the film and the quality of the barrier layers applied. Thus very high barrier effect against gases, such as oxygen and flavors, is

achieved in metallized, biaxially oriented polyester films. A barrier effect against steam is achieved in metallized, biaxially oriented polypropylene films.

The good barrier properties of metallized or oxidically coated films mean that they are used in particular for packaging foodstuffs and luxury foods, for which long storage or transport times create the risk that the packed foodstuffs become spoilt, rancid or lose flavor if there is an inadequate barrier; examples are coffee, snacks containing fats (nuts, chips, etc.) and drinks containing carbon dioxide (in pouches).

If polyester films metallized with an aluminum layer or having an applied oxidic layer are used as packaging material, they are generally a constituent of a multilayer composite film (laminate). Bags produced therefrom can be filled, for example, on a vertical tubular bag forming, filling and sealing machine. The bags are heat-sealed on their inward side (i.e. on the side facing the contents), the heat-sealable layer consisting generally of polyethylene or polypropylene. The composite film here typically has the following structure: polyester layer/aluminum or oxide layer/adhesive layer/heat-sealable layer. If the laminate thickness is from about 50 to 150 μm , the thickness of the metal or oxide layer is only from 10 to 80 nm. Even this very thin layer is sufficiently effective to achieve adequate protection from light and very good barrier properties.

The oxygen barrier or the oxygen transmission is generally measured not on the laminate or the packaging itself, but on the metallized polyester film. To ensure good quality of the foodstuffs or luxury food even after relatively long storage times, the oxygen transmission (identical with permeability) of the metallized film may not be greater than 2 cm^3/m^2 bar d, but in particular not greater than 1.5 cm^3/m^2 bar d. In future, the demands of the packaging industry will head toward still higher barriers, with attempts to achieve permeability values of significantly less than 1.0 cm^3/m^2 bar d for metallized or oxidically coated films.

In the prior art, there is neither sufficient knowledge of the detailed basis for the barrier effect of metallized or oxidically coated polyester films nor of how this may be decisively improved. Variables which are clearly important are the area of the substrate and the type of substrate polymer and its morphology. It is generally assumed that smooth substrate surfaces result in better barrier properties.

In this connection, Weiss et al., in "Thin Solids Films" 204 (1991), p. 203-216, studied the influence of the surface roughness of a substrate layer on the permeability. For this, polyester films were coated with lacquer which contained various concentrations of titanium dioxide particles. In the experiments described, the concentrations of titanium dioxide particles in the lacquer varied from 2 to 20% by weight. Using this method, the surface roughness R_a of the coated substrate surface could be varied from 43 nm (unlacquered and lacquered film, without titanium dioxide) to 124 nm. In his experiments, increasing roughness (increasing proportion of TiO_2) of the lacquered surface resulted in markedly higher oxygen transmissions after metallizing with aluminum. However, the largest step increase in oxygen transmission was seen when the lacquered film (0% by weight) was compared with the unlacquered film, although the surface roughness of the substrate was the same in both cases. The lacquering alone of the film gave a deterioration in the barrier from about 0.43 cm^3/m^2 d bar (plain film) to about 19 cm^3/m^2 d bar (lacquered film). A further uncertainty concerning the transferability of this work to commercial products is created by the fact that the aluminum

layer was applied using a laboratory evaporator. When compared with an industrial metallizer, this method achieves essentially low permeability values, and the influence of the substrate surface on the barrier properties cannot be seen clearly.

Other detailed results of studies on the influence of the substrate surface of polyester films on their barrier properties can be found in the dissertation by H. Utz (Technische Universität München 1995: "Barriereigenschaften aluminumbelagter Kunststoffolien" [Barrier properties of aluminum-metallized plastic films]).

According to the studies by Utz (pp. 66 ff.), there is no direct correlation between the surface roughness (average roughness height R_a) of the PET film and its oxygen barrier. For example, the film for video applications which, with an average roughness height of $R_a=22$ nm, is highlighted as particularly smooth, has, at $1.3 \text{ cm}^3/\text{m}^2$ bar d, an oxygen transmission of $1.2 \text{ cm}^3/\text{m}^2$ bar d comparable with the much rougher PET II film ($R_a=220$ nm).

EP-A-0 124 291 describes a single-layer biaxially oriented polyester film for magnetic recording tape which has the following surface property parameters

- a) the average roughness R_a is from 1 to 16 nm,
- b) the coefficient of friction μk is from 0.01 to 0.20 and
- c) the following relationship exists between R_a and μk

$$0.1 < 10^4 R_a \mu k < 0.31.$$

These properties are created by using TiO_2 particles (anatase) or TiO_2 and CaCO_3 particles in a proportion by weight of, respectively, from 0.1 to 0.5% and from 0.1 to 0.3%. The diameter of the TiO_2 particles is from 0.1 to 0.5 μm . The surface of this film is formed by a large number of elevations/protrusions which obey a distribution such that the graph described by the following relationship

$$\log y = -8.0x + 4.34, y > 10$$

is not intersected. In this equation, x (μm) is a height above a standard level and y is the number of elevations (number/ mm^2) if the elevations are sectioned at a height of x . The distribution of the elevations is determined using standard equipment for measuring roughness. According to this text, although the processing properties of the film are improved, no information is provided on improvement of the gloss, the haze and the barrier properties of the film. There is also no indication of any kind in the text as to how the topography of a film of this type should be adjusted for simultaneous improvement of gloss and oxygen barrier.

EP-A-0 490 665 A1 describes a single-layer biaxially oriented polyester film for magnetic recording tape; the film contains

- a) from 0.05 to 1.0% by weight of ω -alumina having an average particle diameter in the range from 0.02 to 0.3 μm , and
- b) from 0.01 to 1.5% by weight of inert particles of a type other than ω -alumina and having an average particle diameter in the range from 0.1 to 1.5 μm , these particles being larger than the ω -alumina particles.

The surface of this film is formed by a large number of elevations/protrusions which are described by the relationship

$$-11.4x + 4 \log y < -10.0x + 5, x > 0.05 \mu\text{m}$$

In this equation, x (μm) is a height above a standard level and y is the number of elevations (number/ mm^2) if the elevations are sectioned at a height of x . The distribution of

the elevations is measured as in EP-A-0 124 291. This text, like that mentioned above, gives no information concerning improvement of the gloss, the haze and the barrier properties. It also gives no indication of any type as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

The prior art also discloses films which have different roughnesses on their two surfaces (dual surface). These films are suitable in particular for magnetic recording media and essentially have different topographies (e.g. surface A smooth, surface B rough). These texts generally provide means of improving the processing properties of the film but not its optical properties. In particular, however, these texts do not provide any means of improving the barrier properties of the film.

DE-A-16 94 404 describes a layered material having more than one layer of an oriented crystallizable thermoplastic film and in which at least one of the outer layers contains an additive. The additives are customary inert inorganic or organic particles, and in the case of inert particles such as SiO_2 , are added to the outer layers in concentrations of from 1 to 25% by weight, the particle size being from 2 to 20 μm . The layered materials may, for example, be metallized with aluminum for decorative uses or used for magnetic tape. Although this text provides a means to improve the processing properties and the haze of the film, it does not provide a means for improving the gloss and the barrier properties of the film. The text also gives no indication of any type as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

DE-A-22 30 970 describes a magnetic recording medium which is composed of a biaxially oriented polyester film and a thin magnetic metallic layer on the surface A of the polyester film. The film comprises

- a) a coated surface A, which is free from particles and
 - i) is at least 4 μm thick or
 - ii) makes up at least 50% of the thickness of the entire film layer; and
- b) a second layer containing particles and having relatively rough surface and containing
 - i) at least 1% of individual particles of a particular polymer A and
 - ii) at least 1% of individual particles of a particular polymer B.

A disadvantage of the film is that the surface A tends to block, so that the film does not process well. The text does not disclose a means for improving the gloss, the haze and the barrier properties of the film. Once again, there are also no indications of any type in the text as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

EP-B-0 061 769 describes a magnetic recording medium which is formulated from a biaxially oriented polyester film and a thin magnetic metallic layer on the surface A of the polyester film. If desired, there is also a lubricant layer on the other surface B of the polyester film. Features of the film are that the coated surface A

- a) has an average roughness R_a (peak-valley value) of not more than 5 nm (60 nm),
- b) the number of protrusions having a height of from 0.27 to 0.54 μm is from 0 to 0.2 per mm^2 and
- c) is free from protrusions having a height greater than 0.54 μm .

A disadvantage of the film is that the surface A tends to block, so that the film does not process well. The text gives no teaching on improvement of the gloss, the haze and the

barrier properties of the film. There is also no indication of any type in the text as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

EP-B-0 088 635 describes a coextruded biaxially oriented polyester film having at least two layers, of which a layer A consists of thermoplastic resin and a layer B comprises thermoplastic resin and fine particles. The surface roughness R_a of the outer layer on the layer A in the film is less than 5 nm and the outer surface of the layer B is either

- i) a surface having a surface roughness R_a of from 5 to 40 nm and a large number of depressions and a large number of protrusions which are arranged in a particular arrangement or
- ii) a surface which has protrusions formed on a level area and which is covered by a layer C, which consists of a lubricant and has a surface roughness R_a of from 5 to 40 nm.

A disadvantage of this film surface is that it blocks, both with itself and with certain other surfaces (e.g. rubber rolls). The film cannot be processed cost-effectively; in particular during metallizing in vacuo, this film, because of its high tendency to block, tends to tear, and this can cause great cost problems. The film is unsuitable for the purposes of the object to be achieved. In addition, the haze of the film is unsatisfactory.

EP-B-0 502 745 describes a coextruded biaxially oriented polyester film having at least three layers, of which an outer layer A

- a) contains inorganic particles having an average primary particle size D in the range from 1 to 100 nm and satisfying the equation $D < T < 200D$, where T is the thickness of the layer A and
- b) contains particles B having an average primary particle size D1 in the range from 0.3 to 2 μ m, where the primary particle size distribution has a coefficient of variation of not more than 0.6 and
- c) the average primary particle size D of the particles A is smaller than the average primary particle size D1 of the particles B.

The processing behavior of the film, in particular, is improved by applying the teaching of this text. The text does not give any teaching on improving the gloss, the haze or the barrier properties of the film. The text also gives no indication of any type as to how the topography of such a film should be adjusted for simultaneous improvement of gloss and oxygen barrier.

It is also known that the oxygen barrier can be improved by selecting particular polymers for the film serving as substrate (Schricker, G.: Metallisierte Kunststoffolien für höherwertige Verpackungen [Metallized plastic films for high-quality packaging] in: ICI 5th International Metallizing Symposium 1986, Cannes). Polyesters, for example, are particularly suitable, specifically those made from ethylene glycol and terephthalic acid or from ethylene glycol, terephthalic acid and naphthalene-2,6-dicarboxylic acid. Besides these, polyamides, ethylene-vinyl alcohol copolymers (EVOH) and polyvinylidene chloride may also be used with advantage. Thus, for example, U.S. Pat. No. 5,506,014 describes a copolyester made from (a) from 45 to 85 mol % of terephthalic acid, (b) from 10 to 40 mol % of naphthalenedicarboxylic acid and (c) from 5 to 15 mol % of a dicarboxylic acid having from 2 to 8 carbon atoms and (d) ethylene glycol; (the molar percentages are based on the total proportion of dicarboxylic acids). This polyester is claimed to have better barrier properties against gases. It is used, inter alia, for producing bottles or containers and films

of various thicknesses. A disadvantage of the raw materials mentioned is that they are significantly more expensive than polyethylene terephthalate (PET) or are unsuitable and/or not officially permitted for use in the packaging of food-stuffs.

BRIEF SUMMARY OF THE INVENTION

It has been an object of the present invention to provide a coextruded biaxially oriented polyester film which has very good optical properties, i.e. low haze and in particular high gloss, at least of one film surface (termed film surface A below). A surface A of this type would, for example, be particularly advantageous for printing or metallizing. The high gloss of the film is transferred to the print and to the metal layer and thus ensures a particularly brilliant appearance of the packaging. This applies both to reverse printing and also to top printing of the film. In the production of stamping foils, in which the metal layer is transferred from the polyester film onto a carrier made from, for example, board, paper or plastic, the high metallic gloss is retained on the carrier and gives the transfer print an appearance which is particularly effective for presentation purposes.

At the same time, the film should exhibit a high oxygen barrier after metallizing or oxidic coating of the surface A. It should moreover be easy to produce and to process. In summary, the object was to provide a film having the following combination of features:

- high gloss, in particular of the surface A of the film
- low haze
- low oxygen permeability of the film after metallizing or oxidic coating of the surface A of the film
- low coefficients of friction; (no blocking of the film with itself).

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

FIG. 1a is a diagrammatic illustration of surfaces which have a good oxygen barrier.

FIG. 1b is a diagrammatic illustration of surfaces which have a low oxygen barrier.

FIG. 2a is a graph showing the relationship of equation 1 of the present invention.

FIG. 2b is a graph showing the relationship equation 2 of the present invention.

FIG. 3 is a graph showing the relationship of equation 1 of EP 0124 291.

FIG. 4 is an optical microscope photograph of a polyester film.

FIG. 5 is a graph showing distribution of the elevations/protrusions N measured for the outer layer A of the film of Example 1.

DETAILED DESCRIPTION OF THE INVENTION

The gloss of the surface A of the film should be greater than 180 and the haze of the film should be less than 1.5. The finish of the surface of side A should be such that it does not block with itself; (blocking occurs if the coefficient of friction is greater than 1 or if discontinuities arise in the measurement of the variation of frictional force with displacement, when frictional force is measured). The surface of the other side should have a coefficient of friction of less than 0.5.

In the present case, low oxygen permeability means that less than 0.5 cm³ of oxygen per square meter and per day

should diffuse through the metallized or oxidically coated film when it is subjected to 1 bar air pressure.

In its other properties, the film should be at least equivalent to the known packaging films of this type. For example, it should be simple and economic to produce and process well on conventional machinery (i.e. not block, for example).

The object has been achieved by means of a coextruded biaxially oriented polyester film having a base layer B composed to an extent of at least 80% by weight of a thermoplastic polyester and one or more further layers, where at least one outer layer A contains internal and/or inert particles and where this outer layer has a number of elevations/protrusions N per mm^2 of film surface area which is related to their respective heights h and diameters d by the following equations

$$\log N/\text{mm}^2 < A_h - B_h \cdot \log h/\mu\text{m}, \quad (1)$$

$$0.01 \mu\text{m} < h < 10 \mu\text{m}$$

$$A_h = 1.4; B_h = 2.5$$

$$\log N/\text{mm}^2 < A_d - B_d \cdot \log d/\mu\text{m}, \quad (2)$$

$$0.4 \mu\text{m} < d < 10 \mu\text{m}$$

$$A_d = 3.4; B_d = 2.4.$$

For the purposes of the present invention, elevations/protrusions are conical elevations/protrusions which project from the flat film surface.

For the purposes of the invention, internal particles are catalyst residues which remain in the raw material during preparation of the polyester raw materials.

For the purposes of the invention, inert particles are particles which are added to the raw material, for example during its preparation.

To achieve high oxygen barriers in metallized or oxidically coated films according to the object of the invention, the number N of elevations/protrusions per mm^2 of film surface A must, in accordance with the equations (1) and (2), be below a particular numerical value. This numerical value is uniquely defined by the right-hand side of the equations (1) and (2) as a function of the height h and the diameter d of the elevations/protrusions.

Biaxially oriented polyester films which are described by the equations above have a comparatively low number of elevations/protrusions on the layer A which is to be metallized or oxidically coated. The number of elevations/protrusions is in the range $h < 0.5 \mu\text{m}$, in particular $h < 0.4 \mu\text{m}$ and very particularly $h < 0.3 \mu\text{m}$ and is markedly lower than is known in the prior art.

The decisive factors for achieving high oxygen barriers in metallized or oxidically coated films are therefore not low roughness values R_a or R_z , but rather a low density N/mm^2 of elevations/protrusions on the surface A . FIGS. 1a and 1b give a diagrammatic comparison of surfaces which have
a) a good oxygen barrier (low permeability values) according to the present invention (FIG. 1a) and
b) a low oxygen barrier (high permeability values) (FIG. 1b).

The parameter shown is the average roughness R_a , calculated by the following formula

$$R_a = 1/L \cdot \int_0^L f(x) \cdot dx$$

If the density N/mm^2 of the elevations/protrusions is small (FIG. 1a), the barrier in the above sense is good, and

in contrast if the density of the elevations/protrusions is great (FIG. 1b), the barrier in the above sense is then poor. The diagram also shows that in principle the value of R_a has no influence on the barrier properties. A smooth film (e.g. $R_a < 10 \text{ nm}$) here may exhibit a very poor barrier if the number N/mm^2 of elevations/protrusions is greater than calculated by the equations (1) and (2). In this case, the surface/surface layer contains very many fine particles, but these do not contribute significantly to the value of R_a . A surface of this type is not at all suitable for achieving high barrier values. In contrast, film surfaces which have comparatively few elevations/protrusions N per unit of area of the layer A are very suitable for achieving high barrier values. It is of relatively subordinate significance here whether the elevations/protrusions are the result of large particles or of small particles.

While the R_a values have no practical significance for the barrier properties, they do have practical significance for the processing of the novel film. R_a on the film surface A in accordance with the invention and having improved processability is greater than 6 nm, preferably greater than 8 nm and particularly preferably greater than 10 nm.

R_a of the film surface A in accordance with the invention is not greater than 100 nm, advantageously less than 80 nm and particularly advantageously less than 70 nm.

If R_a of the film surface in accordance with the invention is less than 6 nm, the processing, and in particular the reeling, of the film becomes difficult. If R_a of the film surface in accordance with the invention is greater than 100 nm, the abrasion resistance of this surface becomes poorer.

If the number N of elevations per unit of surface on the film layer A which is to be metallized or oxidically coated is greater than the right-hand side of equation (1) or (2), then the oxygen permeability is greater than $0.5 \text{ cm}^3/\text{m}^2 \text{ bar d}$, and this is undesirable for the object of the present invention. The gloss of this film surface (in its unmetallized or uncoated condition) is then moreover no longer sufficiently high to meet the requirements of the object of the present invention.

In a preferred embodiment of the novel film, the constant A_h in equation (1) mentioned above is 1.18, and in a particularly preferred embodiment the value A_h is 1.0. In a likewise preferred embodiment of the novel film, the constant B_h in equation (1) mentioned above is 2.2, and in a particularly preferred embodiment B_h is 2.1.

In a preferred embodiment of the novel film, the constant A_d in equation (2) mentioned above is 3.0, and in a particularly preferred embodiment A_d is 2.6. In a likewise preferred embodiment, the constant B_d of equation (2) mentioned above is 2.3, and in a particularly preferred embodiment B_d is 2.2.

In the preferred and particularly preferred embodiments, the layer A in accordance with the invention has an extremely low number N of elevations/protrusions per unit of surface area. In this case, the metallized or oxidically coated film has a particularly good oxygen barrier. The permeability values of the metallized or oxidically coated film in the preferred embodiment are less than $0.45 \text{ cm}^3/\text{m}^2 \text{ bar d}$, and in the particularly preferred embodiment are less than $0.40 \text{ cm}^3/\text{m}^2 \text{ bar d}$. The gloss of this side likewise then assumes extremely high values. The gloss of this side of the film (in its unmetallized or uncoated condition) in the particularly preferred embodiment is greater than 200.

FIGS. 2a and 2b show, respectively, equations (1) and (2) graphically. When shown using two logarithmic axes, both relationships are straight lines defined by the numerical values given.

Relationships similar to equations (1) and (2) are given in the prior art in the abovementioned texts EP-A-0 124 291 and EP-A-0 490 665. However, as already mentioned, the films claimed in these texts have excellent slip properties as a result of many very small elevations ("... excellent slipperiness of the polyester film of this invention is simultaneously achieved by the presence of the many very minute protrusions (page 7)"; the film contains many inert fine particles ("contains many inert solid fine particles (page 9)"), and this is precisely not the case in the novel films.

In addition, the measurement method used in the prior art is markedly different from that used in this application (cf. description of the measurement method hereinafter. Corresponding to this, the topographies of the film surfaces may also differ, as is apparent on comparing the graphs (FIGS. 2a, 2b and 3) and from the comparative examples. For further illustration of the differences between the measurement methods and of the resultant differences in the topography of the films, FIG. 4 shows an optical microscope photograph (DIC, Differential Interference Contrast) of a polyester film. This photograph, taken using reflected light, shows a pigment-filled polyester film. In the method used here (in accordance with the invention), as described hereinafter, all of the elevations/protrusions are recorded by means of a scanning electron microscope and evaluated using an image analysis method. In contrast, the measurement method of the prior art uses a pin which scans the surface at certain distance intervals. As can be seen in the image, straight line traces are left behind by the pin. The image also shows clearly that with this method

only a few pigment particles are recorded and

the pigment particles are encountered only by chance.

The method of the prior art is thus not reproducible and gives false information.

The comparative examples show quantitatively that, when compared with the prior art, the films in accordance with the present invention have markedly different surface topographies.

The subclaims give preferred embodiments of the invention which are additionally explained below.

In accordance with the invention, the film has at least two layers. The layers which it comprises are then a layer B and the layer A in accordance with the invention. In a preferred embodiment of the invention, the film is built up from three layers and has, on the one side of the layer B (base layer) the layer A and, on the other side of the layer B, another layer C which is made from polyethylene terephthalate and which contains the pigments useful for the production and processing of the film. In this case, the two layers A and C form the outer layers A and C.

In principle, various raw materials may be used for the materials of the various layers. However, it is preferable that the production of the individual layers is based on polyester raw materials.

The base layer B of the film is preferably composed to an extent of at least 90% by weight of a thermoplastic polyester. Polyesters suitable for this are those made from ethylene glycol and terephthalic acid (polyethylene terephthalate, PET), from ethylene glycol and naphthalene-2,6-dicarboxylic acid (polyethylene 2,6-naphthalate, PEN), from 1,4-bis(hydroxymethyl)-cyclohexane and terephthalic acid (poly-1,4-cyclohexanedimethylene terephthalate, PCDT) or from ethylene glycol, naphthalene-2,6-dicarboxylic acid and biphenyl-4,4'-dicarboxylic acid (polyethylene 2,6-naphthalate bibenzoate, PENBB). Particular preference is given to polyesters which are composed to an extent of at least 90 mol %, preferably at least 95 mol

%, of ethylene glycol and terephthalic acid units or of ethylene glycol and naphthalene-2,6-dicarboxylic acid units. The remaining monomer units are derived from other aliphatic, cycloaliphatic or aromatic diols and dicarboxylic acids, which may also be present in the layer A (or the layer C).

Examples of other suitable aliphatic diols are diethylene glycol, triethylene glycol, aliphatic glycols of the formula $\text{HO}-(\text{CH}_2)_n-\text{OH}$, where n is an integer from 3 to 6, (in particular 1,3-propanediol, 1,4-butanediol, 1,5-pentanediol and 1,6-hexanediol), or branched aliphatic glycols having up to 6 carbon atoms. Of the cycloaliphatic diols, cyclohexanediols (in particular 1,4-cyclohexanediol) should be mentioned. Examples of other suitable aromatic diols are those of the formula $\text{HO}-\text{C}_6\text{H}_4-\text{X}-\text{C}_6\text{H}_4-\text{OH}$, where X is $-\text{CH}_2-$, $-\text{C}(\text{CH}_3)_2-$, $-\text{C}(\text{CF}_3)_2-$, $-\text{O}-$, $-\text{S}-$ or $-\text{SO}_2-$. Besides these, bisphenols of the formula $\text{HO}-\text{C}_6\text{H}_4-\text{C}_6\text{H}_4-\text{OH}$ are also very suitable.

Other preferred aromatic dicarboxylic acids are benzenedicarboxylic acids, naphthalenedicarboxylic acids (for example naphthalene-1,4- or -1,6-dicarboxylic acid), biphenyl-x,x'-dicarboxylic acids (in particular biphenyl-4,4'-dicarboxylic acid), diphenylacetylene-x,x'-dicarboxylic acids (in particular diphenylacetylene-4,4'-dicarboxylic acid) and stilbene-x,x'-dicarboxylic acids. Of the cycloaliphatic dicarboxylic acids, mention should be made of cyclohexanedicarboxylic acids (in particular cyclohexane-1,4-dicarboxylic acid). Particularly suitable aliphatic dicarboxylic acids are the C_3 - C_{19} alkanedioic acids, the alkane part of which may be straight-chain or branched.

The polyesters may be prepared by the transesterification process, the starting materials for which are dicarboxylic esters and diols, which are reacted using the customary transesterification catalysts, such as salts of zinc, calcium, lithium, magnesium and manganese. The intermediates are then polycondensed in the presence of widely used polycondensation catalysts, such as antimony trioxide or titanium salts. The preparation may be carried out just as successfully by the direct esterification process in the presence of polycondensation catalysts, starting directly from the dicarboxylic acids and the diols.

Processes which have proven particularly suitable are those in which transesterification catalysts are used with which only a small number of, and/or only small, elevations/protrusions are created on the layer A of the film. Magnesium salts and manganese salts are particularly preferred here. These transesterification catalysts are preferred for preparing the raw material for the base and are particularly preferred for preparing the raw material for the layer A.

Films having the required topography (equations (1) and (2)) are obtained if (in particular for the layer A in accordance with the invention) the raw materials used are prepared using, for example, Mn transesterification catalysts, Mg transesterification catalysts or Ca transesterification catalysts in which the Mn content is in the range from 50 to 200 ppm or the Mg content is in the range from 100 to 300 ppm or the Ca content is in the range from 50 to 300 ppm. This creates internal particles which give films having the surface in accordance with the invention. It is, however, also possible to use other transesterification catalysts.

The concentrations given here for the transesterification catalysts are based on raw materials for the layer A in accordance with the invention in which no additional inert particles are used. In the case of addition of inert particles, it is expedient to reduce the content of internal particles.

In principle, the same polymers may be used for the layer A as for the base layer. Besides these, other materials may

also be present in the layer A, in which case the layer A may preferably be composed of a mixture of polymers, of a copolymer or of a homopolymer which contain ethylene 2,6-naphthalate units and ethylene terephthalate units. Up to 10 mol % of the polymers may be composed of other comonomers (see above).

For the other layer (outer layer C) or for any intermediate layers which may be present, it is possible in principle to use the polymers already described for the base layer and the layer A.

For processing the polymers, it has proven useful to select the polymers for the base layer and the other layer(s) in such a way that the viscosities of the respective polymer melts do not differ excessively. Otherwise it is likely that there will be additional elevations/protrusions, flow disturbances or streaking in the finished film. To describe the viscosity ranges of the two melts, use is made of a modified solution viscosity (SV). For commercially available polyethylene terephthalates which are suitable for producing biaxially oriented films, the SV values are in the range from 600 to 1000. For the purposes of the present invention, to ensure a satisfactory film quality, the SV of the polymers for the layers A or C should be in the range from 500 to 1200, preferably from 550 to 1150, particularly preferably from 600 to 1000. If necessary, a solid phase condensation may be carried out on the respective granules in order to adjust the SV values of the materials as necessary. The SV values of the polymer melts for the base layer and the other layer(s) should not differ by more than 200 units, preferably by not more than 150 units and in particular by not more than 100 units.

The base layer and the other layer(s) may also contain customary additives, such as stabilizers and/or anti-blocking agents. They are expediently added to the polymer or to the polymer mixture before melting takes place. Examples of stabilizers are phosphorus compounds, such as phosphoric acid and phosphoric esters.

Typical anti-blocking agents (also termed pigments in this context) are inorganic and/or organic particles, for example calcium carbonate, amorphous silicic acid, talc, magnesium carbonate, barium carbonate, calcium sulfate, barium sulfate, lithium phosphate, calcium phosphate, magnesium phosphate, alumina, LiF, the calcium, barium, zinc and manganese salts of the dicarboxylic acids used, carbon black, titanium dioxide, kaolin and crosslinked polystyrene particles and crosslinked acrylate particles.

Selected additives may also be mixtures of two or more different anti-blocking agents or mixtures of anti-blocking agents of the same formulation but of different particle size. The particles may be added to the individual layers in the respective advantageous concentrations, e.g. as glycolic dispersion during the polycondensation or via masterbatches during extrusion. Pigment concentrations of from 0 to 5% by weight have proven particularly suitable. A detailed description of the anti-blocking agents which can be used is found, for example, in EP-A-0 602 964.

To fulfill the equations (1) and (2), the layer A in accordance with the invention is generally filled only slightly or not at all with inert pigments. The concentrations of the inert particles in the layer A is from 0 to 0.08% by weight, preferably from 0 to 0.065% by weight, in particular from 0 to 0.05% by weight, and very preferably from 0 to 0.04% by weight, and depends essentially on the size of the particles used. Preferred particles are SiO_2 in colloidal and in chain form. There is no limit in principle on the particle diameters of the particles used. However, to achieve the object of the invention it has proven advantageous to use particles having an average primary particle diameter of less than 60 nm, preferably less than 55 nm and particularly preferably less than 50 nm and/or particles having an average primary particle diameter of greater than 1 μm , preferably greater than 1.5 μm and particularly preferably greater than 2 μm .

If no inert particles are added to the layer A, it should be ensured, for example by a suitable selection and concentration of the catalysts for the transesterification or polymerization reaction, that the precipitates which form can prevent blocking of the surface A with itself.

In selecting the pigments, the pigment diameter and the pigment concentration for the other layer(s) (e.g. for the layer B if the film has two layers or for the base layer B and the outer layer C if the film has three layers) care should be taken that the pigmentation and the layer thicknesses of these layers are selected so that equations (1) and (2) are not contravened.

The pigmentation of the individual layers unrelated to layer A can therefore vary greatly and depends essentially on the structure of the film (layer structure) and the requirements of the film with respect to achievement of other optical properties (haze) and on the behavior in production and processing.

For example, if the film is the preferred three-layer film having the base layer B and the two outer layers A and C, the particle concentration in the base layer B is then preferably lower than in the second outer layer C. The pigmentation in the base layer B should be selected so that it has no lasting effect on the number N of elevations/protrusions in the outer layer in accordance with the invention. In a three-layer film of the type mentioned, the particle concentration in the base layer B will be from 0 to 0.06% by weight, preferably from 0 to 0.04% by weight, in particular from 0 to 0.03% by weight and very preferably from 0 to 0.02% by weight. There is in principle no restriction on the particle diameter of the particles used, but particles having an average diameter of greater than 1 μm are particularly preferred.

If the film is built up from two layers, the layer A making up more than 50%, in particular more than 65% and particularly preferably more than 80% of the total thickness of the film, the layer B can then be much more strongly pigmented (with higher pigment concentration) than in the opposite case in which the thickness of the layer A is comparatively low (<50%).

In a particularly preferred embodiment, the novel polyester film is built up from three layers and then also contains a layer C. The two layers A and C then form the outer layers A and C. The structure, thickness and formulation of the second outer layer C may be selected independently of the outer layer A already present, but the second outer layer may likewise contain the polymers or polymer mixtures already mentioned, which, however, need not be identical with those of the first outer layer. The second outer layer may also contain other well known outer layer polymers. This second outer layer generally contains more pigments (i.e. higher pigment concentrations) than the first outer layer A in accordance with the invention. The pigment concentration in this second outer layer is from 0.02 to 0.4%, advantageously from 0.025 to 0.3%, in particular from 0.03 to 0.2% and very particularly preferably from 0.035 to 0.15%. It depends, for example, on the desired processing behavior of the film. The pigment type(s), the pigment concentration(s) and the particle concentration(s), and also the layer thickness ratios, are preferably selected so that the film has good optical properties and is easy to produce and to process.

It has proven advantageous to describe the ease of production and the ease of processing of the film by means of the following parameters of the side C

- average roughness $R_{a,C}$
- static/sliding friction μ_C of this side with itself and
- the number N_C/mm^2 of elevations/protrusions.

It is advantageous if the film is constructed so that, on this surface C (or B in the case of a film having two layers), which is opposite to the outer layer in accordance with the invention,

13

- a) R_a is from 20 to 100 nm
 b) the static/sliding friction μ_c of this layer with itself is less than 0.5 and
 c) the number N_c/mm^2 of elevations/protrusions is expressed by the equations

$$A_{k2}-B_{k2} \cdot \log h/\mu\text{m} < \log N_c/\text{mm}^2 < A_{k3}-B_{k3} \cdot \log h/\mu\text{m} \quad (3)$$

$$0.01 \mu\text{m} < h < 10 \mu\text{m}$$

$$A_{k2}=-1.000; B_{k2}=3.70$$

$$A_{k3}=2.477; B_{k3}=2.22$$

$$A_{d2}-B_{d2} \cdot \log d/\mu\text{m} < \log N_c/\text{mm}^2 < A_{d3}-B_{d3} \cdot \log d/\mu\text{m} \quad (4)$$

$$0.4 \mu\text{m} < d < 10 \mu\text{m}$$

$$A_{d2}=1.700; B_{d2}=3.86$$

$$A_{d3}=4.700; B_{d3}=2.70$$

In a preferred embodiment, R_a is from 30 to 90 nm and in particular from 35 to 80 nm.

In a preferred embodiment, the static/sliding friction F_c of this layer with itself is less than 0.45 and in particular less than 0.40.

In a preferred embodiment, the constants in equation (3) have the values $A_{k2}=-0.523$, $B_{k2}=3.523$, $A_{k3}=2.300$ and $B_{k3}=2.3$; in a particularly preferred embodiment $A_{k2}=0.00$, $B_{k2}=3.300$, $A_{k3}=2.000$ and $B_{k3}=2.400$, and very particularly preferably $A_{k2}=1.420$, $B_{k2}=2.500$, $A_{k3}=2.000$ and $B_{k3}=3.000$.

In a preferred embodiment, the constants of equation (4) have the values $A_{d2}=2.00$, $B_{d2}=3.630$, $A_{d3}=4.40$ and $B_{d3}=2.70$; in a preferred embodiment $A_{d2}=2.400$, $B_{d2}=3.720$, $A_{d3}=4.000$ and $B_{d3}=2.600$, and very particularly preferably $A_{d2}=3.400$, $B_{d2}=2.400$, $A_{d3}=4.000$ and $B_{d3}=3.300$.

If desired, there may be an intermediate layer between the base layer and the outer layer(s). This also may be composed of the polymers described for the base layers. In a particularly preferred embodiment, it is composed of the polyester used for the base layer. It may also contain the customary additives described. The thickness of the intermediate layer is generally greater than $0.3 \mu\text{m}$ and is preferably in the range from 0.5 to $15 \mu\text{m}$, in particular from 1.0 to $10 \mu\text{m}$ and very particularly preferably from 1.0 to $5 \mu\text{m}$.

In the particularly advantageous embodiment of the novel film, which has three layers, the thickness of the outer layer(s) A (and C) is generally greater than $0.1 \mu\text{m}$ and is generally in the range from 0.2 to $3.0 \mu\text{m}$, advantageously from 0.2 to $2.5 \mu\text{m}$, in particular from 0.3 to $2 \mu\text{m}$ and very particularly preferably from 0.3 to $1.5 \mu\text{m}$; the outer layers A and C here may have the same or different thicknesses.

The total thickness of the novel polyester film can vary within wide limits and depends on the intended application. It is from 4 to $50 \mu\text{m}$, in particular from 5 to $45 \mu\text{m}$, preferably from 6 to $40 \mu\text{m}$, the layer B being a proportion of from 5 to 90% of the total thickness.

To prepare the layers A and C (outer layer(s) A and C), it is expedient to feed pellets of polyethylene terephthalate to one or two extruders. The materials are melted at about 300°C . and extruded.

The polymers for the base layer are expediently fed through another extruder. Any foreign bodies or contamination which may be present can be screened out from the polymer melt before extrusion. The melts are then shaped in a coextrusion die to give flat melt films, and are laminated together. The multilayer film is then drawn off and solidified with the aid of a chill roll and, if desired, other rolls.

The biaxial orientation is generally carried out sequentially. For this, it is preferable to orientate firstly in a longitudinal direction (i.e. in the machine direction) and then in a transverse direction (i.e. perpendicularly to the machine

14

direction). This causes an orientation of the molecular chains. The orientation in a longitudinal direction may be carried out with the aid of two rolls running at different speeds corresponding to the stretching ratio to be achieved. For the transverse orientation, use is generally made of an appropriate tenter frame.

The temperature at which the orientation is carried out can vary over a relatively wide range and depends on the film properties desired. In general, the longitudinal stretching is carried out at from 80 to 130°C ., and the transverse stretching at from 90 to 150°C . The longitudinal stretching ratio is generally in the range from $2.5:1$ to $6:1$, preferably $3:1$ to $5.5:1$. The transverse stretching ratio is generally in the range from $3.0:1$ to $5.0:1$, preferably from $3.5:1$ to $4.5:1$. Before the transverse stretching, one or both film surfaces may be in-line coated by the known processes. The in-line coating may, for example, serve to improve the adhesion of the metallic layer or of any printing ink which may be applied, or else to improve the antistatic or processing behavior.

In the subsequent heat-setting, the film is held for from 0.1 to 10 s at a temperature of from 150 to 250°C . The film is then reeled up in a customary manner.

Before printing or before applying the metallic or oxidic layer on one or both sides, the biaxially oriented and heat-set polyester film may be corona- or flame-treated. The intensity of treatment is selected so that the surface tension of the film is generally greater than 45 mN/m .

Metallic or oxidic layers, if desired, are applied in customary industrial systems. Metallic layers of aluminum are usually produced by conventional metallization (boast method). Besides this, sputtering or electron beam processes in particular have proven successful for oxidic layers. The process parameters of the system when applying the metallic layer or the oxidic layer to the film correspond to the standard conditions. The metallizing of the films is preferably carried out in such a way that the optical density of the metallized film is in the usual range of from about 2.2 to 2.8 . The oxidic layer is applied to the film in such a way that the oxide layer thickness is preferably in the range from 30 to 100 nm . The web speed of the film to be coated is usually from 5 to 20 m/s for all settings of variables.

The film may be coated or corona- or flame-pretreated to establish other desired properties. Typical coatings are those which promote adhesion, are antistatic, improve slip or have release action. These additional coatings may be applied to the film via in-line coating using aqueous dispersions, before the transverse orientation.

If the film is metallized on the layer A in accordance with the invention, the metallic layer is preferably composed of aluminum. However, other materials which can be applied in the form of a thin, cohesive layer are also suitable. Silicon, for example, is particularly suitable and, in contrast to aluminum, gives a transparent barrier layer. The oxidic layer is preferably composed of oxides of elements of the 2nd, 3rd or 4th main group of the Periodic Table, in particular oxides of magnesium, aluminum or silicon. Use is generally made of those metallic or oxidic materials which can be applied at reduced pressure or in vacuo. The thickness of the layer applied is generally from 10 to 100 nm .

It is an advantage of the invention that the production costs of the novel film are comparable with those in the prior art. The other properties of the novel film which are relevant to its processing and use are essentially unchanged or even improved. Besides this, it has been ensured that reclaim can be used during the production of the film. The term "reclaim" as used herein refers to materials including imperfect or discarded film such as edge trim or leftover film that is recycled and reprocessed with fresh polyethylene terephthalate pellets in production of the novel film. Reclaim can

be used in a concentration of from 20 to 50% by weight, based on the total weight of the film, without any significant adverse effect on the physical properties of the film.

The film has excellent suitability for packaging of food-stuffs and luxury foods which are sensitive to light and/or air. It is particularly suitable for producing vacuum packs for coffee, in particular ground coffee.

In summary, the novel film has high gloss, in particular high gloss of the surface A of the film, and low haze. The film is also an excellent oxygen barrier after it has been metallized or coated with oxidic materials on the surface A of the film. In addition, it has good reeling and processing behavior.

The gloss of the surface A of the film is greater than 180. In a preferred embodiment, the gloss of this side is greater than 190 and in a particularly preferred embodiment greater than 200. This surface of the film is therefore suitable in particular for printing or metallizing. The high gloss of the film is transferred to the print or the applied metallic layer and thus imparts to the film the effective presentational appearance which is wanted.

The haze of the film is less than 1.5. In a preferred embodiment, the haze of the film is less than 1.3 and in a particularly preferred embodiment less than 1.0. Its low haze makes the film suitable not only for packaging but also, for example, for reprographic or "glazing" applications.

The oxygen transmission of the film is less than $0.5 \text{ g m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$, preferably less than $0.45 \text{ g m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ and particularly preferably less than $0.4 \text{ g m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$ after it has been metallized or coated with oxidic materials on the surface A of the film.

No blocking occurs on the surface A of the film. The coefficient of friction on the side opposite to the side A is less than 0.5. In a preferred embodiment, the coefficient of friction of that part of the film is less than 0.45 and in a particularly preferred embodiment less than 0.4.

Table 1 below shows once again the most important film properties in accordance with the invention.

TABLE 1

	range according to the invention	preferred	particularly preferred	unit	measurement method
Gloss, side A (20° angle of measurement) ¹⁾	>180	>190	>200		DIN 67 530
Haze ¹⁾	<1.5	<1.3	<1.0	%	ASTM-D 1003-52
Oxygen permeability of the metallized or oxidically coated film	<0.5	<0.45	<0.40	$\text{g m}^{-2} \text{ d}^{-1} \text{ bar}^{-1}$	DIN 53 380, Part 3
Coefficient of friction: Side A with itself Side C and/or side B, respectively, with itself	no blocking <0.5	<0.45	<0.40		DIN 53 375
Average roughness R_a Side C and/or side B	20-100	30-90	35-80	nm	DIN 4768, with a cut-off of 0.25 nm

¹⁾Measured on the unmetallized film

The following methods were used to determine parameters for the raw materials and the films:

(1) Optical Density

The Macbeth TD-904 Densitometer from Macbeth (Division of Kollmorgen Instruments Corp.) was used to measure the optical density. The optical density is defined as $OD = -\lg I/I_0$, where I is the intensity of the incident light, I_0 is the intensity of the transmitted light and I/I_0 is the transmittance.

(2) Oxygen Barrier

The oxygen barrier of the metallized films was measured using an OX-TRAN 2/20 from Mocon Modern Controls (USA) in accordance with DIN 53 380, Part 3.

(3) SV

The SV (solution viscosity) was determined by dissolving a specimen of polyester in a solvent (dichloroacetic acid). The viscosity of this solution and that of the pure solvent were measured in an Ubbelohde viscometer. The quotient was determined from the two values, 1,000 was subtracted from this, and this value multiplied by 1000. The result was the SV.

(4) Coefficient of Friction

The coefficient of friction was determined according to DIN 53 375, 14 days after production. Blocking takes place if the coefficient of friction is greater than 1 or if discontinuities occur in the plot of frictional force against displacement during measurement of the frictional force.

(5) Surface Tension

The surface tension was determined using the "ink method" (DIN 53 364).

(6) Haze

The haze of the film was measured according to ASTM-D 1003-52. The Hölz haze was determined by a method based on ASTM-D 1003-52, but in order to utilize the most effective measurement range, measurements were made on four pieces of film laid one on top of the other, and a 1° slit diaphragm was used instead of a 4° pinhole.

(7) Gloss

Gloss was measured according to DIN 67 530. The reflectance was measured as a characteristic optical value for a film surface. Based on the standards ASTM-D 523-78 and ISO 2813, the angle of incidence was set at 20° or 60°. A beam of light hits the flat test surface at the set angle of incidence and is reflected and/or scattered thereby. A proportional electrical variable is displayed, representing the light beams hitting the photoelectric detector. The value measured is dimensionless and must be stated together with the angle of incidence.

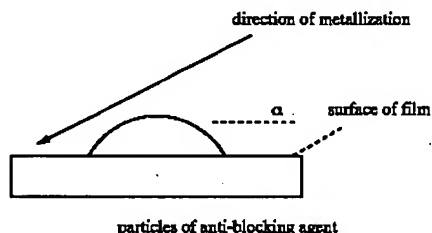
(8) Determination of the Particle Sizes on Film Surfaces

A scanning electron microscope (e.g. DSM 982 Gemini, Leo GmbH (Zeiss)) together with an image analysis system was used to determine the size distribution of particles of anti-blocking agent (particle size distribution) on film surfaces. The magnifications selected in all cases were 1700 times.

For these measurements, specimens of film are placed flat on a specimen holder. These are then metallized obliquely at an angle α with a thin metallic layer (e.g. of silver). α here

17

is the angle between the surface of the specimen and the direction of diffusion of the metal vapor. The anti-blocking agent particles throw a shadow in this oblique metallization. Since the shadows are not yet electrically conductive, the specimen can then be further metallized with a second metal (e.g. gold), the metal vapor here impacting vertically onto the surface of the specimen.



Scanning electron microscope (SEM) images are taken of specimen surfaces prepared in this way. The shadows of the particles of anti-blocking agent are visible because of the contrast between materials. The specimen is oriented in the SEM so that the shadows run parallel to the lower edge of the image (x direction). SEM images are taken with this setting and transferred to an image analysis system. This image analysis system is used to make precise measurements of the lengths of the shadows (in the x direction) and their maximum extent in the y direction (parallel to the vertical edge of the image).

The diameter D of the particles of anti-blocking agent at the surface level of the specimen is equal to the maximum extent of the shadows d in the y direction. The height of the particles of anti-blocking agent, measured from the film surface, can be calculated from the angle α of metallization and the length L of the shadows, given knowledge of the magnification V selected for the SEM image:

$$h = (\tan(\alpha) \cdot L) / V$$

So as to achieve a sufficiently high level of statistical reliability, precise measurements are made of a few thousand particles of anti-blocking agent. With the aid of known statistical methods, frequency distributions are then produced for the diameters and heights of the particles. The class interval selected for this is 0.2 μm for the particle diameter d and 0.05 μm for the particle height h.

(9) Roughness

The roughness R_a of the film was determined according to DIN 4768 with a cut-off of 0.25 nm.

EXAMPLE 1

Polyethylene terephthalate chips (prepared via the transesterification process using Mn as transesterification catalyst; Mn concentration: 100 ppm) were dried at 160° C. to a residual moisture of less than 50 ppm and fed to the extruder for the base layer B.

In addition, polyethylene terephthalate chips (prepared via the transesterification process using Mn as transesterification catalyst; Mn concentration: 100 ppm) which have been pigmented as shown in Table 2 were likewise dried at 160° C. to a residual moisture of less than 50 ppm and fed to the respective extruders for the outer layers A and C.

A transparent film having three layers, ABC structure and a total thickness of 12 μm was produced by coextrusion followed by stepwise orientation in longitudinal and transverse directions. The thickness of the respective layers is given in Table 2.

18

Outer layer A is a mixture of:

94.0% by weight of polyethylene terephthalate having an SV of 800
6.00% by weight of masterbatch made from 99.0% by weight of polyethylene terephthalate (SV of 800) and 0.5% by weight of Sylobloc @ 44 H (colloidal SiO_2 from Grace) and 0.5% by weight of Aerosil @ TT 600 (chain SiO_2 from Degussa)
Base layer B:

100.0% by weight of polyethylene terephthalate having an SV of 800

Outer layer C is a mixture of:

84.0% by weight of polyethylene terephthalate having an SV of 800
16.0% by weight of masterbatch made from 99.0% by weight of polyethylene terephthalate and 0.5% by weight of Sylobloc 44 H (Grace) and 0.5% by weight of Aerosil TT 600 (Degussa)

The production conditions for the individual process steps were:

Extrusion:	Temperatures Layer A:	300° C.
	Layer B:	300° C.
	Layer C:	300° C.
	Die gap width	1 mm
Longitudinal stretching:	Temperature of the take-off roll	30° C.
	Temperature:	80–125° C.
Transverse stretching:	Longitudinal stretching ratio:	4.0
	Temperature:	80–135° C.
	Transverse stretching ratio:	4.0
Setting:	Temperature:	230° C.
Duration:		3 s

The film has very good optical properties and good processing behavior (cf. Table 3).

After the film had been produced (in this Example 1 and in all examples below) it was metallized on the side A with aluminum in vacuo in an industrial metallizer. The coating speed was 8 m/s and the optical density was 2.6.

The film exhibited the required high oxygen barrier. The structure of the film and the properties achieved in films produced in this way are presented in Tables 2 and 3. FIG. 5 shows the distributions for the elevations/protrusions N measured for the outer layer A using the method as described hereinabove.

EXAMPLE 2

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 1. Compared with Example 1, only the outer layer A was changed.

Outer layer A is a mixture of:

98.0% by weight of polyethylene terephthalate having an SV of 800

-continued

Outer layer A is a mixture of:	
2.00% by weight of	masterbatch made from 99.0% by weight of polyethylene terephthalate (SV of 800) and 0.5% by weight of Sylobloc 44 H (Grace) and 0.5% by weight of Aerosil TT 600 (Degussa)

The process conditions selected for all layers were as in Example 1.

EXAMPLE 3

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 1. Compared with Example 1, only the outer layer A was changed.

Outer layer A is a mixture of:	
99.0% by weight of	polyethylene terephthalate having an SV of 800
1.00% by weight of	masterbatch made from 99.0% by weight of polyethylene terephthalate (SV of 800) and 0.5% by weight of Sylobloc 44 H (Grace) and 0.5% by weight of Aerosil TT 600 (Degussa)

The process conditions selected for all layers were as in Example 1.

EXAMPLE 4

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 1. Compared with Example 1, only the outer layer A was changed.

Outer layer A is a mixture of:	
98.0% by weight of	polyethylene terephthalate having an SV of 800
2.00% by weight of	masterbatch made from 99.75% by weight of polyethylene terephthalate (SV of 800) and 0.25% by weight of Aerosil TT 600 (Degussa)

The process conditions selected for all layers were as in Example 1.

EXAMPLE 5

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 1. Compared with Example 1, only the outer layer A was changed.

Outer layer A is a mixture of:	
98.0% by weight of	polyethylene terephthalate having an SV of 800
2.00% by weight of	masterbatch made from 99.75% by weight of polyethylene terephthalate (SV of 800) and 0.25% by weight of Sylobloc 44 H (Grace)

The process conditions selected for all layers were as in Example 1.

EXAMPLE 6

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 1. Compared with Example 1, only the outer layer A was changed.

Outer layer A is a mixture of:
100.0% by weight of polyethylene terephthalate having an SV of 800

The polyester raw material for the outer layer A was prepared via the transesterification process using Ca as transesterification catalyst, the Ca concentration being 200 ppm.

The process conditions selected for all layers were as in Example 1. Although no extra inert pigments were added to the outer layer A and the base layer B, the surface of the outer layer A contains elevations/protrusions. These stem, inter alia, from the transesterification catalyst precipitates and variations in the processes of raw material preparation and film production.

EXAMPLE 7

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 1. Compared with Example 1, the outer layers A and C were changed.

Outer layer A is a mixture of:	
98.0% by weight of	polyethylene terephthalate having an SV of 800
2.00% by weight of	masterbatch made from 99.0% by weight of polyethylene terephthalate (SV of 800) and 1.0% by weight of Sylobloc 44 H (Grace)

Outer layer C is a mixture of:	
90.0% by weight of	polyethylene terephthalate having an SV of 800
10.0% by weight of	masterbatch made from 99.0% by weight of polyethylene terephthalate (SV of 800) and 1.0% by weight of Sylobloc 44 H (Grace)

EXAMPLE 8

A transparent film having three layers, ABC structure and a total thickness of 12 μm was prepared by coextrusion

21

followed by stepwise orientation in longitudinal and transverse directions, in a similar manner to that of Example 7. Compared with Example 1, the outer layer A was changed.

Outer layer A is a mixture of:	
91.0% by weight of	polyethylene terephthalate having an SV of 800
1.00% by weight of	masterbatch made from 99.0% by weight of polyethylene terephthalate (SV of 800) and 1.0% by weight of Sylobloc 44 H (Grace)

COMPARATIVE EXAMPLE 1

Example 1 of EP-A-0 514 129 was repeated. The gloss of the film is unsatisfactory. The metallized version in a thickness of 12 μm did not have the required barrier values.

COMPARATIVE EXAMPLE 2

Example 1 of EP-A-0 604 057 was repeated. The gloss of the film is unsatisfactory. The metallized version in a thickness of 12 μm did not have the required barrier values. In

22

addition, the film does not present a cost-effective solution with respect to use in the packaging market.

COMPARATIVE EXAMPLE 3

Example 1 of EP-A-0 124 291 was repeated.

COMPARATIVE EXAMPLE 4

Example 1 of EP-A-0 490 665 was repeated.

COMPARATIVE EXAMPLE 5

Example 1 of DE-A-16 94 404 was repeated.

COMPARATIVE EXAMPLE 6

Example 15 of EP-A-0 061 769 was repeated.

COMPARATIVE EXAMPLE 7

Example 1 of EP-B-0 088 635 was repeated.

TABLE 2

Example	Film thickness μm	Film structure	Layer thicknesses μm A B C	Pigments in the layers			Average pigment diameter μm		Pigment concentrations		
				A	B	C	B		B		
							A	C	A	ppm	C
Example 1	12	ABC	1.0/9.5/1.5	Sylobloc 44 H	none	Sylobloc 44 H	2.5	2.5	300	0	800
				Aerosil TT 600	none	Aerosil TT 600	0.04	0.04	300	0	800
Example 2	12	ABC	1.0/9.5/1.5	Sylobloc 44 H	none	Sylobloc 44 H	2.5	2.5	100	0	800
				Aerosil TT 600	none	Aerosil TT 600	0.04	0.04	100	0	800
Example 3	12	ABC	1.0/9.5/1.5	Sylobloc 44 H	none	Sylobloc 44 H	2.5	2.5	50	0	800
				Aerosil TT 600	none	Aerosil TT 600	0.04	0.04	50	0	800
Example 4	12	ABC	1.0/9.5/1.5	Aerosil TT 600	none	Sylobloc 44 H	2.5	2.5	0	0	800
				Aerosil TT 600	none	Aerosil TT 600	0.04	0.04	50	0	800
Example 5	12	ABC	1.0/9.5/1.5	Sylobloc 44 H	none	Sylobloc 44 H	2.5	2.5	50	0	800
				Aerosil TT 600	none	Aerosil TT 600	0.04	0.04	0	0	800
Example 6	12	ABC	1.0/9.5/1.5	—	none	Sylobloc 44 H	2.5	2.5	0	0	800
				—	none	Aerosil TT 600	0.04	0.04	0	0	800
Example 7	12	ABC	1.0/9.5/1.5	Sylobloc 44 H	none	Sylobloc 44 H	2.5	2.5	200	0	1000
Example 8	12	ABC	1.0/9.5/1.5	Sylobloc 44 H	none	Sylobloc 44 H	2.5	2.5	100	0	1000
Comp. Ex. 1	75	ABA	4/68/4	Glass beads	none	Glass beads	2.7	2.7	300	0	300
				Aerosil OX 50	none	Aerosil OX 50	0.04	0.04	1200	0	1200
Comp. Ex. 2	60	ABA	1.5/63/1.5	Tbaperl 130	none	Tbaperl 130	3.0	3.0	600	0	600
Comp. Ex. 3	10	A	—	—	Anatase - TiO_2	—	0.4	—	—	1500	—
Comp. Ex. 4	14	A	—	—	$\theta\text{-Al}_2\text{O}_3$	—	0.2	—	—	3000	—
Comp. Ex. 5	25	ABA	2.5/20/2.5	kaolin	—	kaolin	4	4	1250	—	1250
Comp. Ex. 6	15	AB	7.5/7.5	zinc acetate	—	kaolin +	—	—	230	5000+	—
				antimony trioxide	—	lubricant as coating	—	—	400	solution	—
Comp. Ex. 7	12	AB	6/6	—	—	internal particles	—	1.5	—	2000	—

TABLE 3

Example	Constants for the height distribution of the particles		thickness distribution of the particles		Oxygen barrier cm^3/m^2	Coefficient of friction		Roughness R_a		Gloss ^D		Haze ^D %	Processing behavior
	A_h	B_h	A_d	B_d		μk		Side C					
	Side A/Side C	Side A/Side C	Side A/Side C	Side A/Side C	bar d	A/A	C/C	Side A	nm	Side A	Side C		
Example 1	1.3/2.45	1.5/2.6	3.3/2.4	3.5/3.1	0.45	0.45	0.4	25	60	185	180	1.7	very good
Example 2	1.25/2.4	1.5/2.6	3.2/2.3	3.5/3.1	0.43	0.5	0.4	20	60	190	180	1.3	very good
Example 3	1.2/2.2	1.5/2.6	3.18/2.3	3.5/3.1	0.43	0.55	0.4	18	60	190	180	1.3	very good
Example 4	1.18/2.2	1.5/2.6	3.1/2.3	3.5/3.1	0.42	0.65	0.4	12	60	195	180	1.2	good

TABLE 3-continued

Example	Constants for the height distribution of the particles		thickness distribution of the particles		Oxygen barrier cm ³ /m ² bar d	Coefficient of friction		Roughness R _a		Gloss ^D		Haze ^D %	Processing behavior
	A _h B _h		A _d B _d			μ k		Side C					
	Side A/Side C		Side A/Side C			A/A	C/C	Side A	nm	Side A	Side C		
Example 5	1.16/2.2	1.5/2.6	3.1/2.3	3.5/3.1	0.41	0.65	0.4	15	60	195	180	1.1	good
Example 6	1.12/2.15	1.5/2.6	3.0/2.3	3.5/3.1	0.40	0.65	0.4	11	60	200	180	1.1	good
Example 7	1.25/2.4	1.65/2.7	3.3/2.3	2.8/3.1	0.42	0.55	0.3	25	70	190	160	1.3	good
Example 8	1.2/2.3	1.65/2.7	3.1/2.3	2.8/3.1	0.42	0.6	0.3	18	70	195	160	1.1	good
Comp. Ex. 1					1.3	0.46	0.46	35	35	160	160	1.3	good
Comp. Ex. 2					0.6	0.26	0.26	55	55	165	165	0.6	moderate
Comp. Ex. 3					2.4		0.15		10		130	7	good
Comp. Ex. 4					3.1		0.2		20		120	9	good
Comp. Ex. 5					1.8		0.4		75		160	1.5	good
Comp. Ex. 6					0.8	blocked	0.4	3.6	15	165	130	9	bad
Comp. Ex. 7					0.45	blocked	0.3	3	14	180	150	5	very bad

^DMeasured on the metallized film

Side A: metallized outer layer

Side C: unmetallized outer layer

What is claimed is:

1. A biaxially oriented, coextruded polyester film having a base layer B composed to an extent of at least 80% by weight of a thermoplastic polyester and one or more further layers, where at least one outer layer A contains internal and/or inert particles and where this outer layer has a number of elevations/protrusions N per mm² of film surface area which is related to their respective heights h and diameters d by the following equations

$$\log N/\text{mm}^2 < A_h - B_h \cdot \log h/\mu\text{m},$$

$$0.01 \mu\text{m} < h < 10 \mu\text{m}$$

$$A_h = 1.4; B_h = 2.5$$

$$\log N/\text{mm}^2 < A_d - B_d \cdot \log d/\mu\text{m},$$

$$0.4 \mu\text{m} < d < 10 \mu\text{m}$$

$$A_d = 3.4; B_d = 2.4.$$

2. A polyester film as claimed in claim 1, wherein the layer A contains less than 0.08% by weight of an inert filler.

3. A polyester film as claimed in claim 1, wherein the film is a metallized or oxidically coated film having an oxygen transmission of $\leq 0.5 \text{ cm}^3/\text{m}^2 \text{ bar d}$.

4. A polyester film as claimed in claim 3, wherein the metallized or oxidically coated film has an oxygen transmission of $\leq 0.45 \text{ cm}^3/\text{m}^2 \text{ bar d}$.

5. A polyester film as claimed in claim 1, wherein the layer A has a glass transition temperature higher than that of the base layer B.

6. A polyester film is claimed in claim 1, wherein the outer layer A has a thickness of from 0.1 to 3.0 μm .

7. A polyester film as claimed in claim 1, wherein the film is built up from two layers and is composed of the base layer B and an outer layer A.

8. A polyester film as claimed in claim 1, which is built up from three layers and is composed of an outer layer A, the base layer B and a second outer layer C which is applied to the side of the base layer B opposite to that of the outer layer A.

9. A polyester film as claimed in claim 8, wherein the outer layer C is pigmented.

10. A polyester film as claimed in claim 8, wherein the outer layer A is pigmented.

11. A polyester film as claimed in claim 8, wherein at least one outer layer is in-line coated.

12. A polyester film as claimed in claim 1, wherein the outer layer A is pigmented.

13. A polyester film as claimed in claim 1, wherein at least one outer layer is in-line coated.

14. A process for producing a biaxially oriented, multi-layer polyester film as claimed in claim 1, in which polyester melts corresponding to the formulations of the outer and base layers are fed to a coextrusion die, extruded from this die onto a cooling roll and the resultant prefilm is then biaxially oriented and heat-set, wherein at least one outer layer has a number of elevations/protrusions N per mm² of film surface area which is related to their respective heights h and diameters d by the following equations

$$\log N/\text{mm}^2 < A_h - B_h \cdot \log h/\mu\text{m}, \quad (1)$$

$$0.01 \mu\text{m} < h < 10 \mu\text{m}$$

$$A_h = 1.4; B_h = 2.5$$

$$\log N/\text{mm}^2 < A_d - B_d \cdot \log d/\mu\text{m}, \quad (2)$$

$$0.4 \mu\text{m} < d < 10 \mu\text{m}$$

$$A_d = 3.4; B_d = 2.4.$$

15. A process for producing a biaxially oriented, multi-layer polyester film, as claimed in claim 14, in which reclaim is introduced to the coextrusion die in a concentration of from 10 to 50% by weight, based on the total weight of the film.

16. A method for packaging of foodstuffs and luxury foods which comprises packaging said foodstuffs and luxury foods in a polyester film as claimed in claim 1.

17. A method for the production of hot-stamping foils which comprises producing said hot-stamping foils from a polyester film as claimed in claim 1.

* * * * *

UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 6,528,144 B2
DATED : March 4, 2003
INVENTOR(S) : Herbert Peiffer et al.

Page 1 of 1

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

Title page,

Item [57], **ABSTRACT,**

Line 10, " $\log N/\text{mm}^2 < A_{h-Bh} * \log h/\mu\text{m}$, $0.01 \mu\text{m} < h < 10 \mu\text{m}$ " should read -- $\log N/\text{mm}^2 < A_n-B_h * \log h/\mu\text{m}$, $0.01 \mu\text{m} < h < 10 \mu\text{m}$ --.

Line 12, " $\log N/\text{mm}^2 < A_{d-Bd} * \log d/\mu\text{m}$, $0.4 \mu\text{m} < d < 10 \mu\text{m}$ " should read -- $\log N/\text{mm}^2 < A_d-B_d * \log d/\mu\text{m}$, $0.4 \mu\text{m} < d < 10 \mu\text{m}$ --.

Column 24,

Line 43, " $\log N/\text{mm}^2 < A_{h-Bh} * \log h/\mu\text{m}$," should read -- $\log N/\text{mm}^2 < A_h-B_h * \log h/\mu\text{m}$, --.

Line 49, " $\log N/\text{mm}^2 < A_{d-Bh} * \log d/\mu\text{m}$," should read -- $\log N/\text{mm}^2 < A_d-B_d * \log d/\mu\text{m}$, --.

Signed and Sealed this

Eighth Day of July, 2003



JAMES E. ROGAN
Director of the United States Patent and Trademark Office